On the Correct Form of the Nonlinear Optical Susceptibility in Strongly-Driven Semiconductor Quantum Dots

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SCOPE OF THIS WORK: In recent years, significant attention has been given to nonlinear optical phenomena, such as nonlinear optical absorption and dispersion, derived from a strongly-driven quantum transition of semiconductor quantum dots. An important issue that is widely studied is the effect of probe electromagnetic field intensity on the total (nonlinear) absorption coefficient and on the total (nonlinear) index of refraction. Some of the effects that have been proposed is the bleaching effect, optical gain and slow light. The most common and widely used methodology for studying this problem is derived from density matrix equations using perturbation theory and includes first-order (linear) as well as third-order (nonlinear) terms. In this work, we revisit the problem of the total absorption coefficient and the total index of refraction in a symmetric semiconductor quantum dot structure under a strong probe field excitation. We use the two-level model, solve the relevant density matrix equations under the rotating wave approximation and under steady state conditions, and obtain the correct form of the nonlinear optical susceptibility that is then used for the derivation of the formulae of the total absorption coefficient and the total index of refraction under the interaction with a strong probe field. Then, for the specific quantum dot system we compare the results of the total absorption coefficient and the total index of refraction for the two main methodologies (the perturbation result and our result) for different electromagnetic field intensities.

NONLINEAR OPTICAL SUSCEPTIBILITY PERTURBATION RESULT

$$\chi(\omega,I)=\chi^{(1)}(\omega)+\chi^{(3)}(\omega)I,$$

$$\chi^{(1)}(\omega)=\frac{N_e\mu^2T_2}{\varepsilon_0\hbar}\,\frac{i-(\omega-\omega_0)T_2}{1+(\omega-\omega_0)^2T_2^2},$$

$$\chi^{(3)}(\omega)=-\frac{2N_e\mu^4T_1T_2^2}{\varepsilon_0^2\hbar^3n_bc}\,\frac{i-(\omega-\omega_0)T_2}{\left[1+(\omega-\omega_0)^2T_2^2\right]^2}.$$

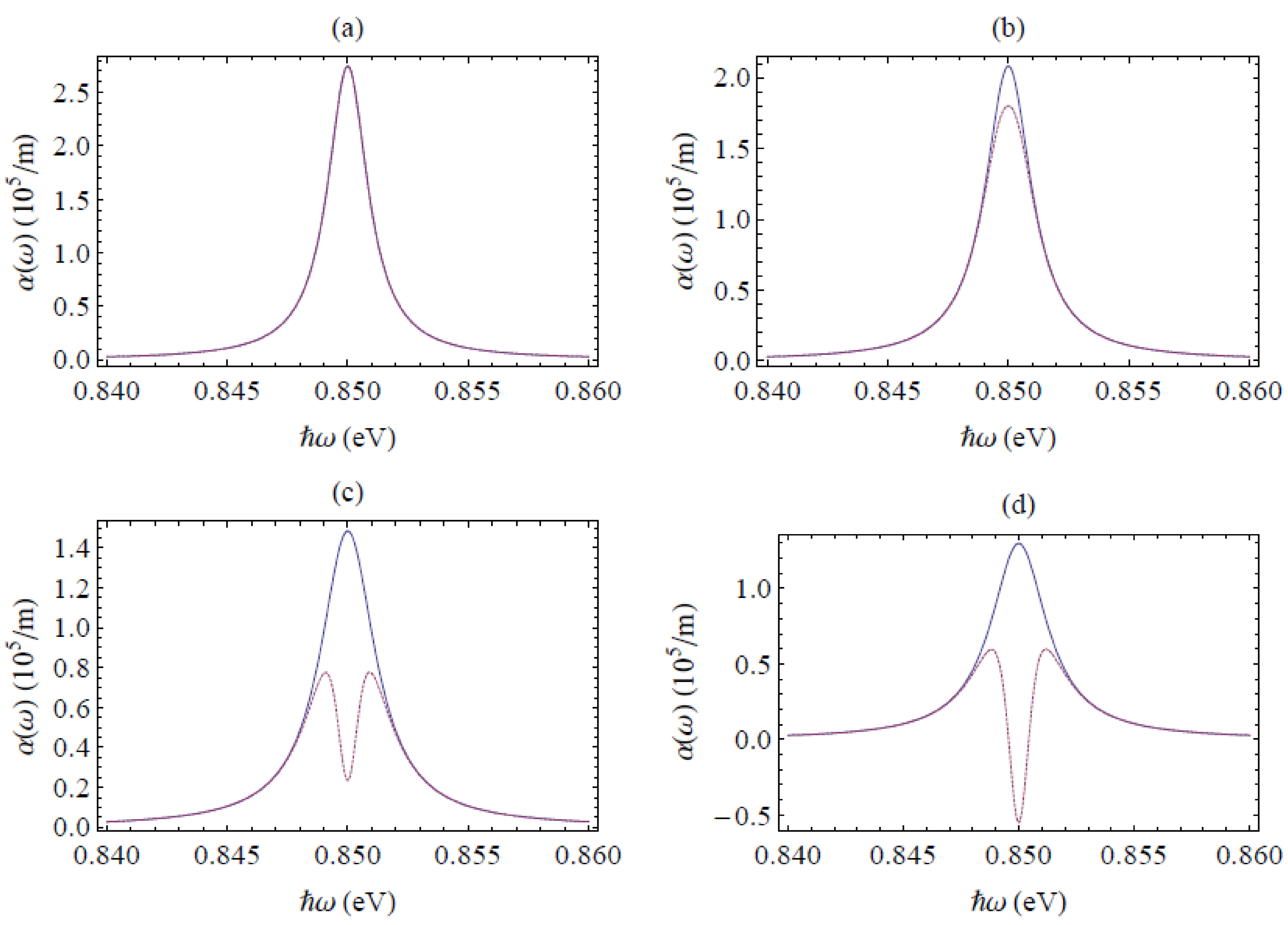
$$\alpha(\omega)=\frac{\omega}{n_bc}\mathrm{Im}(\chi),$$

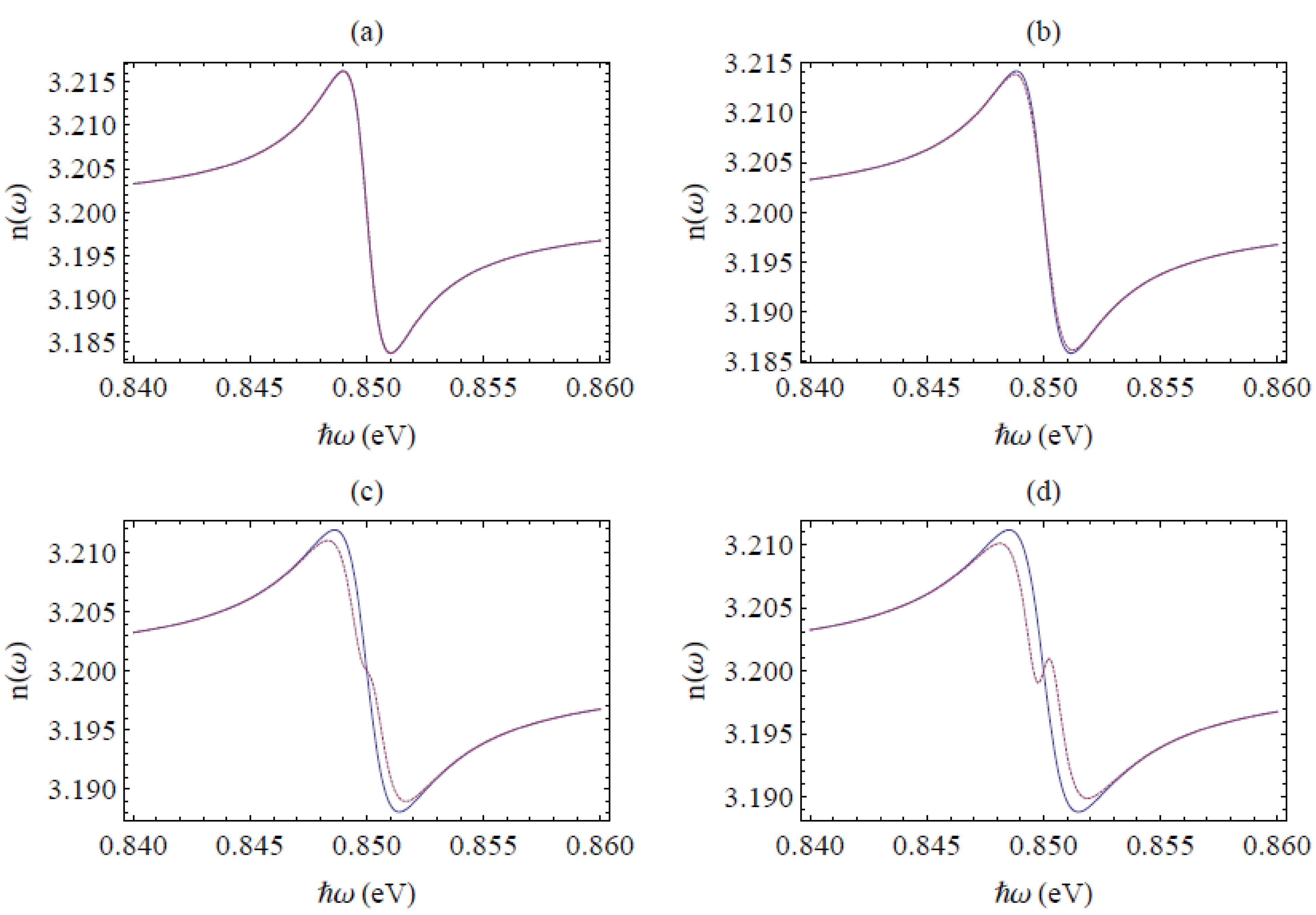
$$n(\omega)=\mathrm{Re}\left(\sqrt{n_b^2+\chi}\right).$$

NONLINEAR OPTICAL SUSCEPTIBILITY OF OUR WORK

$$\chi(\omega,I)=\frac{N_e\mu^2T_2}{\varepsilon_0\hbar}\,\frac{i-(\omega-\omega_0)T_2}{1+(\omega-\omega_0)^2T_2^2+\dfrac{2\mu^2T_1T_2}{cn_b\varepsilon_0\hbar^2}I},$$

- I is the intensity and ω is the frequency of the electromagnetic field that is applied to the quantum dot. Also, ω_0 is the frequency difference and μ the electric dipole matrix element of the two states of the quantum dot that contribute in the dynamics. The relaxation processes in the quantum dot are described phenomenologically by the population decay time T_1 and the dephasing time T_2. In addition, N_e is the volume electron density and n_b the index of refraction of the quantum dot material.





QUANTUM DOT STRUCTURE UNDER STUDY: We consider an InGaAs/InGaAsP quantum dot with disk-like shape and lateral parabolic confinement potential. We derive the electronic structure of the quantum dot system by solving numerically the time-independent Schrödinger equation, under the effective mass approximation, using the potential morphing method and obtain the energy difference and the electric dipole matrix element for the relevant transitions of the quantum dot system. For this system we take $T_1 = 1.5$ ps, $T_2 = 1.5$ ps, $n_b = 3.2$, $N_e = 3\times10^{22}$ m^{-3}.

FIGURE 1 (Left): The total absorption coefficient $\alpha(\omega)$ for the transition from the ground state to the first excited state of the quantum dot for different values of the applied electromagnetic field intensity: (a) $I = 4\times10^8$ W/m^2, (b) $I = 4\times10^9$ W/m^2, (c) $I = 10^{10}$ W/m^2 and (d) $I = 1.3\times10^{10}$ W/m^2. We display with solid blue curves our results and with dotted magenta curves the results of the perturbation theory model.

FIGURE 2 (Left): The total index of refraction $n(\omega)$ for the transition from the ground state to the first excited state of the quantum dot for different values of the applied electromagnetic field intensity: (a) $I = 4\times10^8$ W/m^2, (b) $I = 4\times10^9$ W/m^2, (c) $I = 10^{10}$ W/m^2 and (d) $I = 1.3\times10^{10}$ W/m^2. We display with solid blue curves our results and with dotted magenta curves the results of the perturbation theory model.

DISCUSSION: Both models give the same results for low intensities [Figs. 1(a) and 2(a)]. The phenomena that are commonly found for large electromagnetic field intensities: (i) the creation of a strong dip in the absorption spectrum near or at resonance (the so-called bleaching effect) [Fig. 1(c)], (ii) negative absorption near or at resonance that leads to optical gain [Fig. 1(d)], and (iii) the change of the slope of the total index of refraction near or at resonance from negative to positive [Fig. 2(d)], which changes the behavior of the system from fast to slow light, only appear in the perturbation result (dotted magenta curves) and are artifacts of the perturbation model due to failure of the order of perturbation theory considered. The increase of the applied field intensity leads only to saturation effects to the total absorption coefficient and the total index of refraction, as our model predicts (solid blue curves).

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